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Published in:
International Journal of Advanced Manufacturing Technology

Link to article, DOI:
10.1007/s00170-009-2507-8

Publication date:
2010

Document Version
Publisher's PDF, also known as Version of record

Int J Adv Manuf Technol
DOI 10.1007/s00170-009-2507-8

ORIGINAL ARTICLE

Experimental investigation of the factors influencing the polymer–polymer bond strength during two-component injection moulding

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Received: 21 July 2009 / Accepted: 22 December 2009

Abstract Two-component injection moulding is a commercially important manufacturing process and a key technology for combining different material properties in a single plastic product. It is also one of most industrially adaptive process chain for manufacturing so-called moulded interconnect devices (MIDs). Many fascinating applications of two-component or multi-component polymer parts are restricted due to the weak interfacial adhesion of the polymers. A thorough understanding of the factors that influence the bond strength of polymers is necessary for multi-component polymer processing. This paper investigates the effects of the process conditions and geometrical factors on the bond strength of two-component polymer parts and identifies the factors which can effectively control the adhesion between two polymers. The effects of environmental conditions on the bond strength are also investigated. Investigation shows that melt and mould temperatures are vital process parameters that influence the bond strength. Besides this, surface roughness of the first-shot part and environmental factors like moisture have profound influence on the bonding of the two materials. The selections of materials and environmental conditions were done based on the suitability of MID production, but the results could be useful for two-component polymer processing for a wide range of industrial applications. The results and discussion presented in this paper are only valid for the two-component plastic parts moulded by over moulding in cavity-transfer process.

Keywords Two-component injection moulding · Bond strength · Process parameters · Polymer

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1 Introduction

Two-component (2K) injection moulding is an industrial process to combine two different polymers in a single device. For example, in MIDs, a plateable and non-plateable plastic is combined for selective metallization and to create electrical infrastructures [1]. The challenging task for the two-component moulding is to find a material pair which fulfils the diverse requirements for the engineering application and at the same time has a reasonably good bond between the two polymers in the pair [2]. This paper makes a thorough investigation to understand the factors affecting the polymer–polymer adhesion and sorts out important parameters which could be used as tuning factors for bond strength. The effects of manufacturing process conditions, materials parameters, part geometry, thermal history and environmental conditions were studied and results are presented. Furthermore, the investigation identifies couples of material pairs which could be used for high-performance 2K products as well as for MIDs.

2 Two-component injection moulding

In two-component injection moulding process, two different polymers or the same polymer with two different colours are combined in a single product. Based on the manufacturing process, two-component injection moulding

Published online: 05 February 2010

is divided in sub groups like co-injection moulding, sequential injection moulding and over moulding. The first two types of injection moulding are done in the same machine and mould. Two-component over moulding is done by cavity-transfer process in a single mould–machine system or in two different machines and moulds. For this investigation, two-component parts were produced by the cavity-transfer process in a single mould–machine system.

3 Polymer–polymer adhesion

Adhesion between two polymer materials is a complex phenomenon .The adhesion between two reactive polymers is governed by chemical reaction between the molecules of two polymers at the interface and the strength achieved by this process is extremely high [3]. In case of the same grade of polymer, the adhesion is governed by intermolecular diffusion at the interface. If the material pair is held together under favourable pressure and temperature conditions, the molecules start to diffuse at the interface from either side and after a certain time the interface disappears and the adhesion strength obtained by this process is the same as the strength of material [3]. But the mechanism of polymer adhesion becomes complex when two different polymers interact at the interface, and the degree of adhesion is governed by the entanglement between the molecules of two polymers at the interface [3].

4 Experimental

4.1 Specimen

For the experiments, an ISO-recommended tensile test specimen was chosen as reference geometry. The final two-component test specimens were moulded by cavity-transfer process in a standard injection moulding machine. During the first step of injection moulding, the first-shot part was produced (it was cut into two pieces to the required length after moulding) and in the second step, this was over-moulded using a different plastic material. Figure 1 shows the geometry of the test specimen. The reason for making the second-shot part longer than the first-shot part was to avoid a too-low shot volume in the second-shot injection moulding.

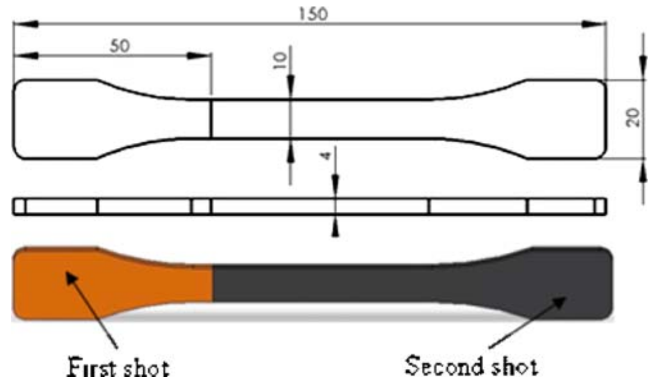


Fig. 1 Geometry of the two-component test specimen

5 Materials and methods

Thermoplastic materials for these experiments were selected on the basis of their use in MID applications and for hearing aid components. Attention was also paid to the most commonly used thermoplastics for engineering applications and to the possibility of metallization. Table 1 contains the list of the materials selected for the experiments. POM and LCP materials were excluded from the experiments because of the weak interfacial adhesion found

Table 1 List of the plastic materials used for the experiments

Name	Trade name	Grade	Manufacturer	Type of crystallinity	T_g	T_m	Process parameters (T_{mo}, T_{melt}, P_{injec}, V_{injec})
Polyetheretherketone (PEEK)	Victrex	150GL30	Victrex	Semi crystalline	153	343	180°C, 380°C, 2210 bar, 112 mm/s
Polyetherimide (PEI)	Ultem	2312	GE	Amorphous	217	282	160°C, 390°C, 2210 bar, 112 mm/s
Polyetherimide (PEI)	Ultem	1000	GE	Amorphous	217	282	160°C, 390°C, 2210 bar, 112 mm/s
Polycarbonate (PC)	Lexan	500R	GE	Amorphous	141	230	90°C, 300°C, 2210 bar, 112 mm/s
Polyphenyleneether blend (PPE+HIPS)	Noryl	GFN1520V	GE	Amorphous	144	225	180°C, 380°C, 2210 bar, 112 mm/s
Polystyrene (PS)	Polystyrol	143E	BASF	Amorphous	88	135	110°C, 280°C, 2210 bar, 112 mm/s
Polystyrene (PS)	Polystyrol	158 K	BASF	Amorphous	100	140	60°C, 230°C, 2210 bar, 112 mm/s
Acrylonitrile-butadiene-styrene (ABS)	Terluran	997VE	BASF	Amorphous	110	170	60°C, 250°C, 2210 bar, 112 mm/s

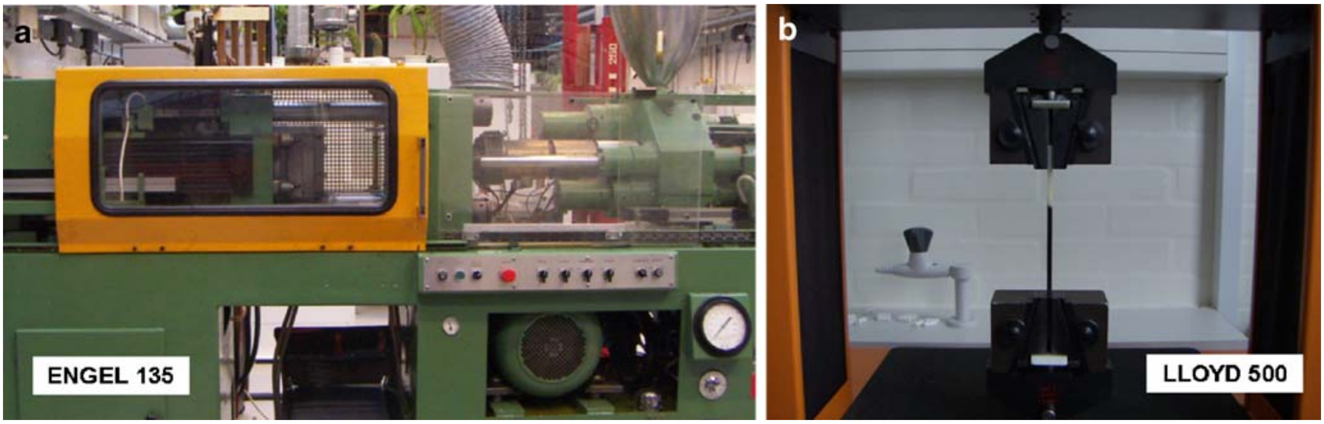
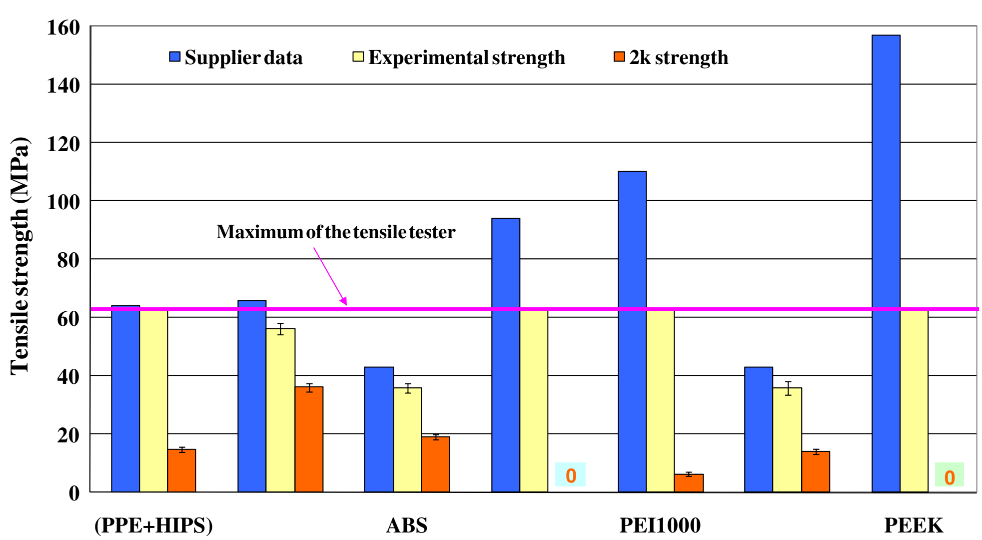


Fig. 2 **a** Injection moulding machine and **b** tensile tester used for the experiments

Fig. 3 Injection moulded 2K test specimen (PEI-first shot and PEEK-second shot)

Fig. 4 Comparative tensile test results of different plastic materials in 1K and 2K (The *error bars* indicate the maximum deviation of a single data point from the average value calculated from five data points)



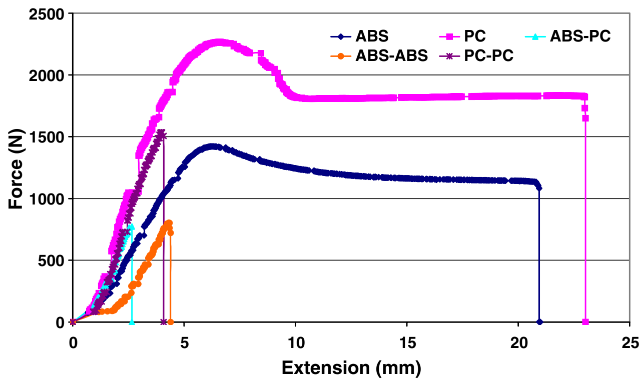


Fig. 5 Comparative tensile test plots of ABS, PC and ABS-PC combinations

in a previous investigation [4]. The listed process parameters are used both in first- and second shot for each material unless otherwise is mentioned and all the parameters correspond to the machine set values.

The injection moulding machine used for the experiment was a modified Engel 135 machine (Fig. 2a). The tensile tester used to determine the bond strength of two polymers was LLOYD 500 produced by Lloyd Instruments (Fig. 2b), and the maximum capacity of the machine was 2,500 N. All the tensile tests were performed at 5 mm/min test speed. Figure 3 shows a finished 2K moulded test specimen made by PEI and PEEK.

5.1 Plan for experimental investigations

Single component and two-component test specimens were produced by injection moulding considering the following investigational plan. The reason to choose different material pair for different test is their suitability to the test conditions.

1. Comparative investigation of tensile strength among supplier data, experimental values and two-component bond strength.
2. Investigation on the effects of injection parameters on polymer–polymer bond strength (ABS-PC combination)
3. Investigation on the effects of interface temperature on the polymer–polymer bond strength (PEEK-PEI, PEEK-PC and PEI-PC combinations)
4. Investigation on the effects of surface roughness and part geometry on the bond strength of two different polymers (ABS-PC combination)
5. Effects of glass fibre in the materials on the bond strength [PS-(PPE+HIPS) and PS-PC combinations]
6. Investigation on the effects of environmental factors on bond strength (ABS-PC, PC-PEI and PEI-PEEK combinations)

6 Results and discussion

6.1 Tensile test results

For the first test, specimens were injection moulded as one shot part (1K filling the entire cavity in one step) and as two-shot part (same material in both the shots). Injection parameters used were the recommended process parameters for the materials and were constant both for 1K and 2K parts. Figure 4 shows the comparative strength of the suppliers' specified value, experimentally determined strength and the strength of two-shot part with the same material. Every result is an average of five experiments. In most of the cases, the

Fig. 6 Bond strength of two-component test specimen (two different polymers in the pair)

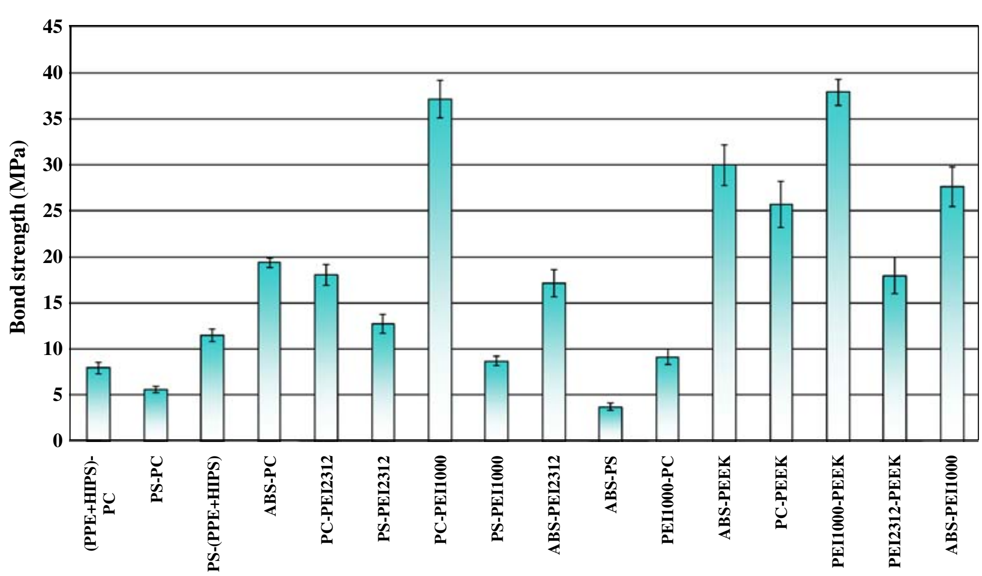


Table 2 Injection moulding process variables used in the experiments

Melt temperature (°C)	Mould temperature (°C)	Injection speed (mm/s)	Injection pressure (bar)	Holding pressure (bar)	Holding time (s)	Cooling time (s)
270	30	48	780	130	5	5
300	60	80	1560	325	10	10
330	90	112	2210	520	15	20

experimental tensile strength was approximately 15% lower than the supplier's specified values. By two-shot injection moulding 30–100% of the strength was lost. The best case was PC–PC which only lost 32% of the 1K strength.

Figure 5 shows the tensile test plots of ABS and PC moulded tensile test specimens made in 1K and 2K combinations. In all cases of 2K moulding, the failure is a so-called "brittle failure" (see Fig. 5, there is no ductile zone in stress–strain curve in case of 2K combinations). On the other hand, both PC and ABS behave as ductile materials in the 1K test.

Figure 6 contains the test results from 2K parts made with two different polymer materials. The highest strength was obtained in the case of PEI-PEEK moulding (37 MPa), also reported in reference [6] PEI also adheres strongly with PC. ABS has reasonably good adhesion with many other polymers.

6.2 Effects of injection parameters

To investigate the effects of injection moulding parameters, ABS inlay parts were over-moulded with PC using different injection parameters. The following injection parameters were studied:

- Melt temperature
- Mould temperature
- Injection speed
- Injection pressure
- Holding pressure
- Holding time
- Cooling time

Table 2 summarises the ranges of different process parameters for the experiments. For the second-shot moulding, a recommended set of process parameters were chosen (texts in the grey background in Table 2). To investigate the effect of any of the above parameters, the values for the rest of the parameters were set to the recommended values. For moulding first-shot ABS parts, the recommended process parameters from Table 1 were used. The reason to use ABS-PC material combinations for this experiment is the extensive use of ABS-PC for industrial 2K application and also the reasonable good adhesion between the material pair.

Figure 7 presents the effects of the each individual parameter on the bond strength of two-shot moulded polymers. The melt temperature and mould temperature are clearly the most important parameters for bond strength. Higher mould and melt temperature increase the interface temperature of the inlay polymer part and the second-shot melt. Higher interface temperature facilitates the melting of the inlay part at the interface so that the two polymers can melt together. Higher interface temperature also increases molecular interdiffusion and entanglement rate at the bond interface.

Injection speed and injection pressure are mutually dependent parameters. Changing one parameter may affect the other one. Still it is clear from the experiment that increasing injection speed and pressure have positive effect on the bonding of two materials. Injection speed affects the interfacial adhesion as the higher injection speed increases the melt temperature due to the shear heating and decreases the melt viscosity. Injection speed and pressure also influence the mechanical locking of two materials so that they can have stronger bonding. Similarly holding pressure increases the mechanical locking of the second-shot polymer melt at the interface and makes the interface stronger. The experimental results show that holding and cooling time affects the bond strength when they are set to an insufficient value and any further increment of these two parameters than the required values does not have any positive effect on the bond strength.

6.3 Interface temperature

Several interesting pairs of polymers were found which had significant difference in bond strength depending on the shot sequence of the two plastics. For example the PEEK-PEI combination had poor bond strength when PEEK was moulded first and PEI was injected in the second shot (4.2 MPa). The same material pair was much stronger simply by reversing the shot sequence (36.9 MPa). To understand this phenomenon, the interface temperature of the inlay part and second-shot melt was calculated according to the following formula (taken from Ref. [5]; Table 3).

$$Ti = \frac{b1T1 + b2T2}{b1 + b2}, \; b = \sqrt{k\rho Cp}$$

The material data were edited from the Moldflow database, CES EduPack 2006 and also from suppliers' data sheet for the second-shot melt and for first-shot solid inlay. The amorphous melting point was calculated by 1/2(glass

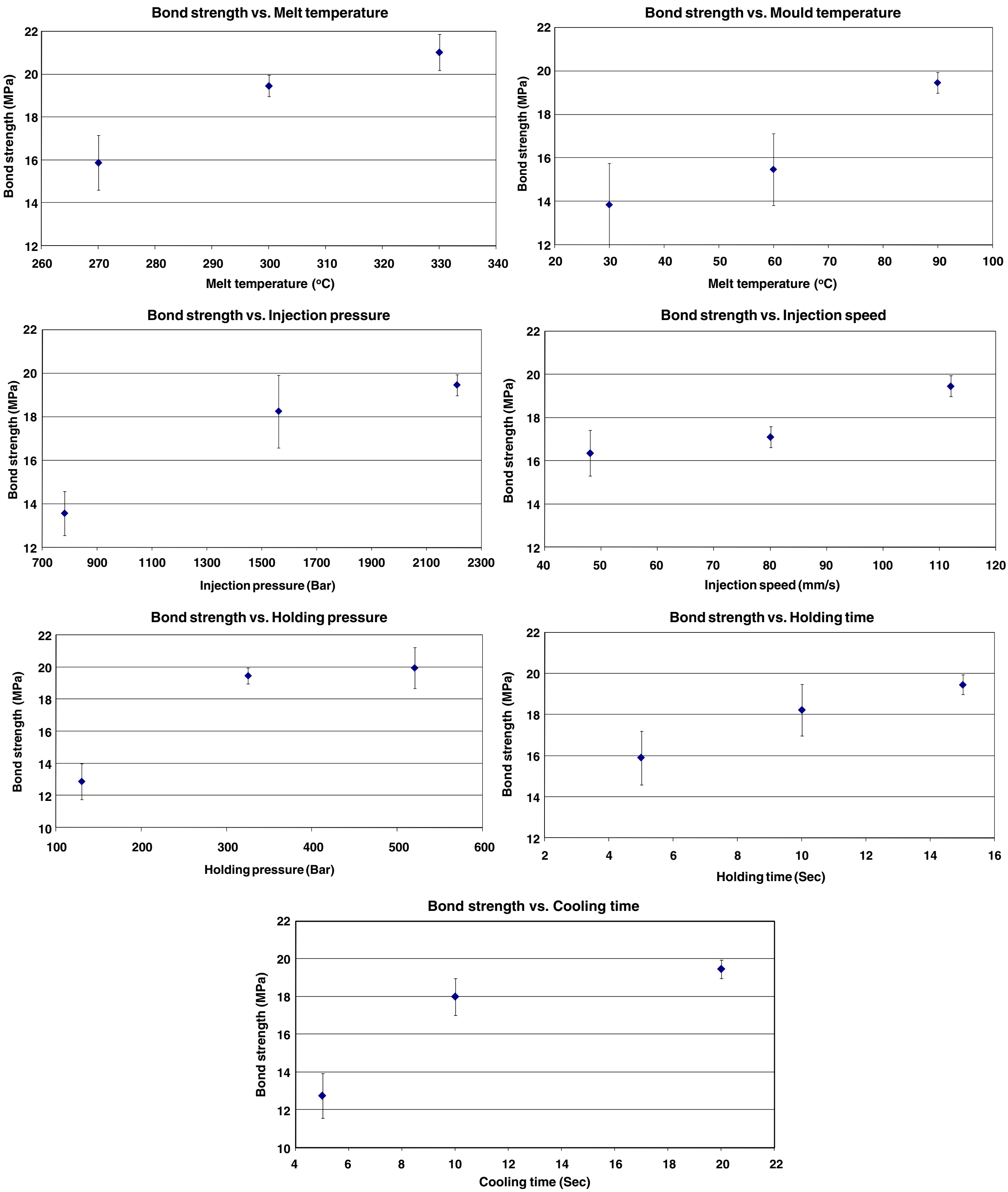


Fig. 7 Effects of different process parameters on polymer–polymer bond strength

Table 3 Effects of interface temperature on bond strength

Shot sequence	Bond strength (MPa)	Interface temperature T_i (°C)	Melting point of the inlay part (°C)
PEEK-PEI	4.2	231	343
PEI-PEEK	36.9	297	282
PEEK-PC	0	200	343
PC-PEEK	26.2	291	225
PEI-PC	9.4	206	282
PC-PEI	37	247	225

T_i interface temperature, b_1 thermal effusivity of the inlay part, b_2 thermal effusivity of the melt, T_1 mould temperature, T_2 second-shot melt temperature, K material thermal conductivity, ρ material density, C_p specific heat capacity of material

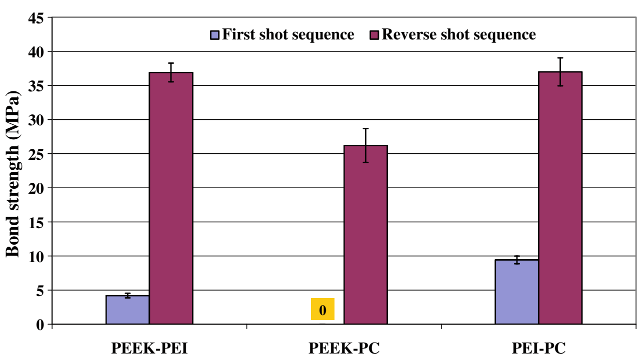


Fig. 8 Comparative bond strength of reverse shot sequence

transition temperature+possible lower process temperature). Possible lower process temperature is the lowest recommended melt temperature for injection moulding given by the material manufacturer. In the first case of PEEK-PEI combination, the calculated interface temperature was 231.7°C and the melting point of inlay PEEK was 343°C. In PEI-PEEK combinations the interface temperature was 297°C and the melting point of the inlay part was 282.5°C and that explains why in the first case bond strength is much lower than in the second case. Figure 8 presents the bond strength of few material pairs which have significant difference in reverse shot sequence and Fig. 9 shows the effects of interface temperature on the bond strength.

6.4 Effects of surface roughness

To find the influence of the interface roughness on the bond strength of two-component parts, three different roughnesses were intentionally generated at the interface surface of the ABS inlay parts by grinding. Three different rough surfaces had S_a values (average roughness values) of 0.59, 1.69 and 3.38 µm, respectively. The measurements were made by a UBM Laser Profilometer. Samples with predefined roughnesses were over-moulded in the second shot by PC. The tensile test results showed a big influence of the interfacial roughness on the bond strength (Fig. 9a). From the plot, it is clear that in a certain range the interfacial roughness can make dramatic change in the adhesion. The rough surface increases the mechanical interlocking of the melt on the inlay surface. It also increases the interfacial contact area of the two polymers. Increased surface area affects the bonding in two possible ways: increases the heat transfer rate from the melt to the solid part and provides a bigger area for mechanical interactions. Besides the mechanical locking, the rough surface facilitates the

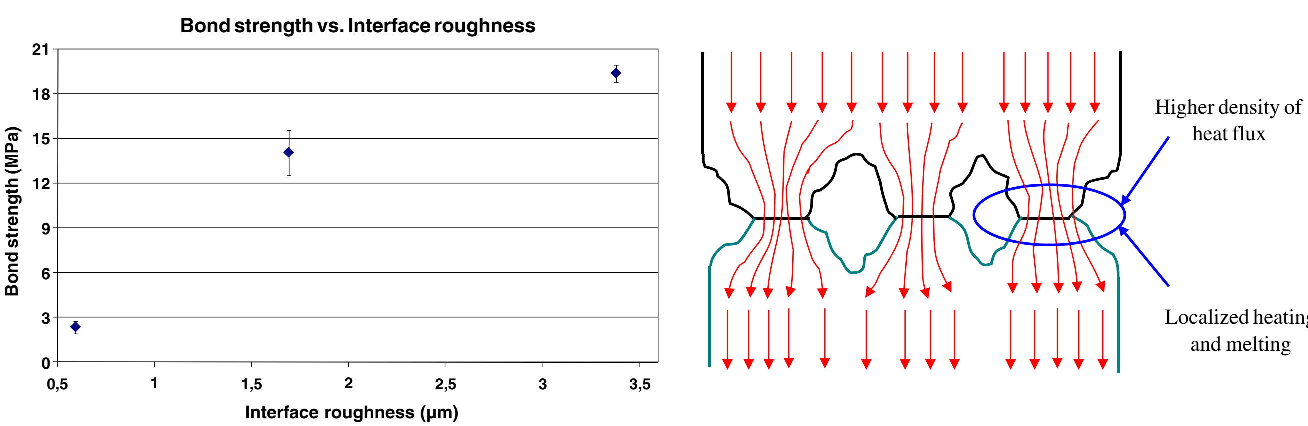


Fig. 9 **a** Bond strength vs. interface roughness. **b** Effects of roughness on localised melting

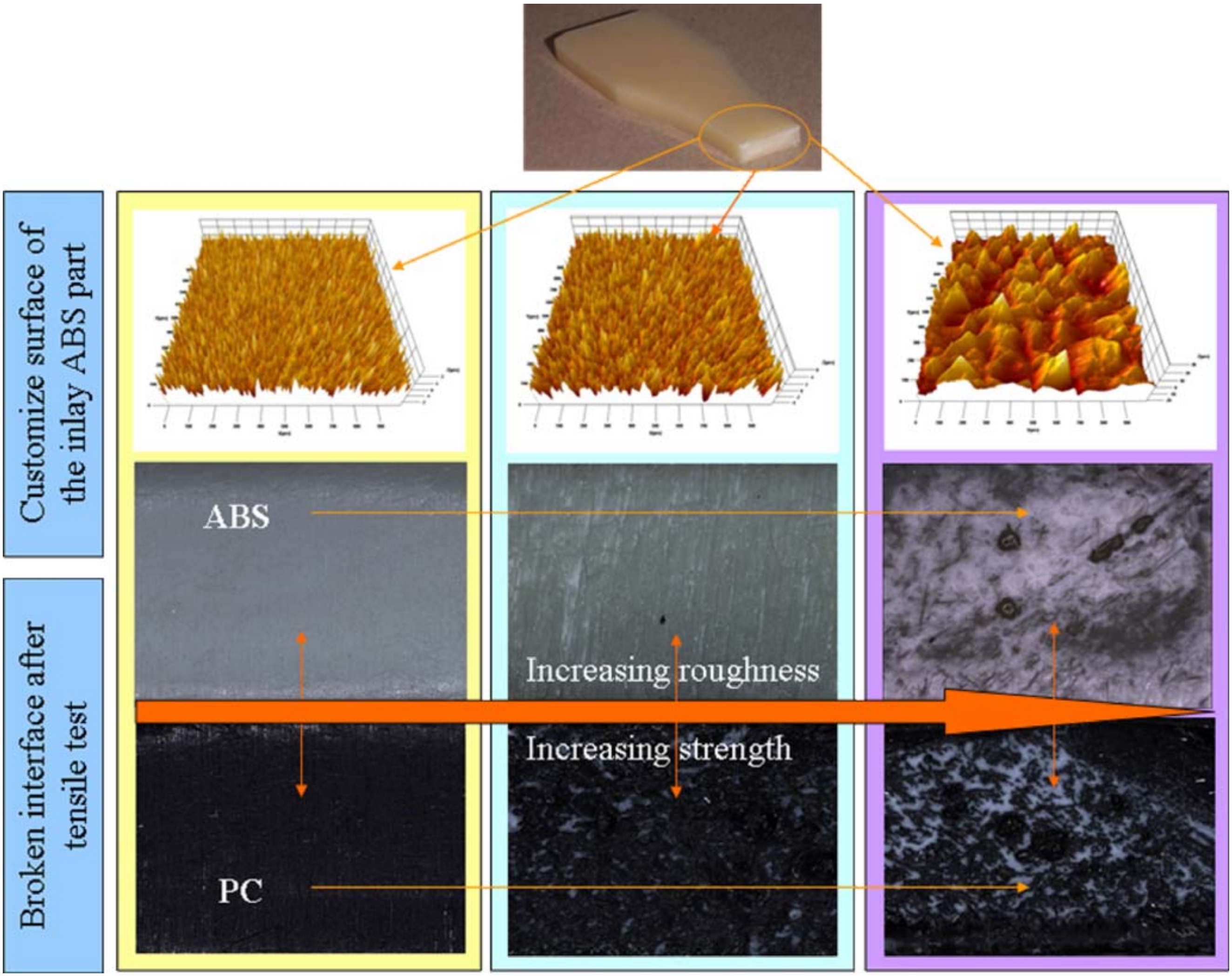


Fig. 10 Effects of interface roughness on polymer–polymer adhesion with the increased roughness increased amount of traces of one material is visible on the other material at the broken surface

localised melting of the inlay part. The upper hills of the rough surface act as an extended heat transfer elements and can easily be melted with the second-shot melt. Figure 9b shows how the roughness peeks concentrate the heat flux and creates localised melting zone. Figure 10 shows the effects of roughness on the interfacial adhesion of ABS-PC material combination. Some other broken interfaces of other material combinations can be viewed in Fig. 11 and it is observable from the failure sites that in case of good adhesion more traces of one material is visible on the other material.

6.5 Effects of glass fibres on polymer–polymer bond strength

Glass fibre influences the tensile strength of polymer significantly. The first plot of Fig. 12 shows the effects of glass fibre on the tensile properties of single component plastic part. Test specimens were moulded with PS 158K with 30% and 0% glass fibre. To test the effect of using glass fibre fillers on the adhesion of two-component plastic part, the first-shot parts were produced using PS158K without glass fibres and with 30% glass fibres. In the second shot, inlay parts were over-moulded using PC and (PPE+HIPS). The tensile tests reveal almost no effects of the glass fibres on the bond strength. The right plot of Fig. 11 shows the test results.

6.6 Effects of environmental factors

The influence of environmental factors on the mechanical and chemical performances of plastics is a well-studied area. With the growing application of two-component injection moulding, the environmental effect on polymer–polymer bond strength is of huge interest. When it comes to the point of two-shot moulded MIDs, the environmental

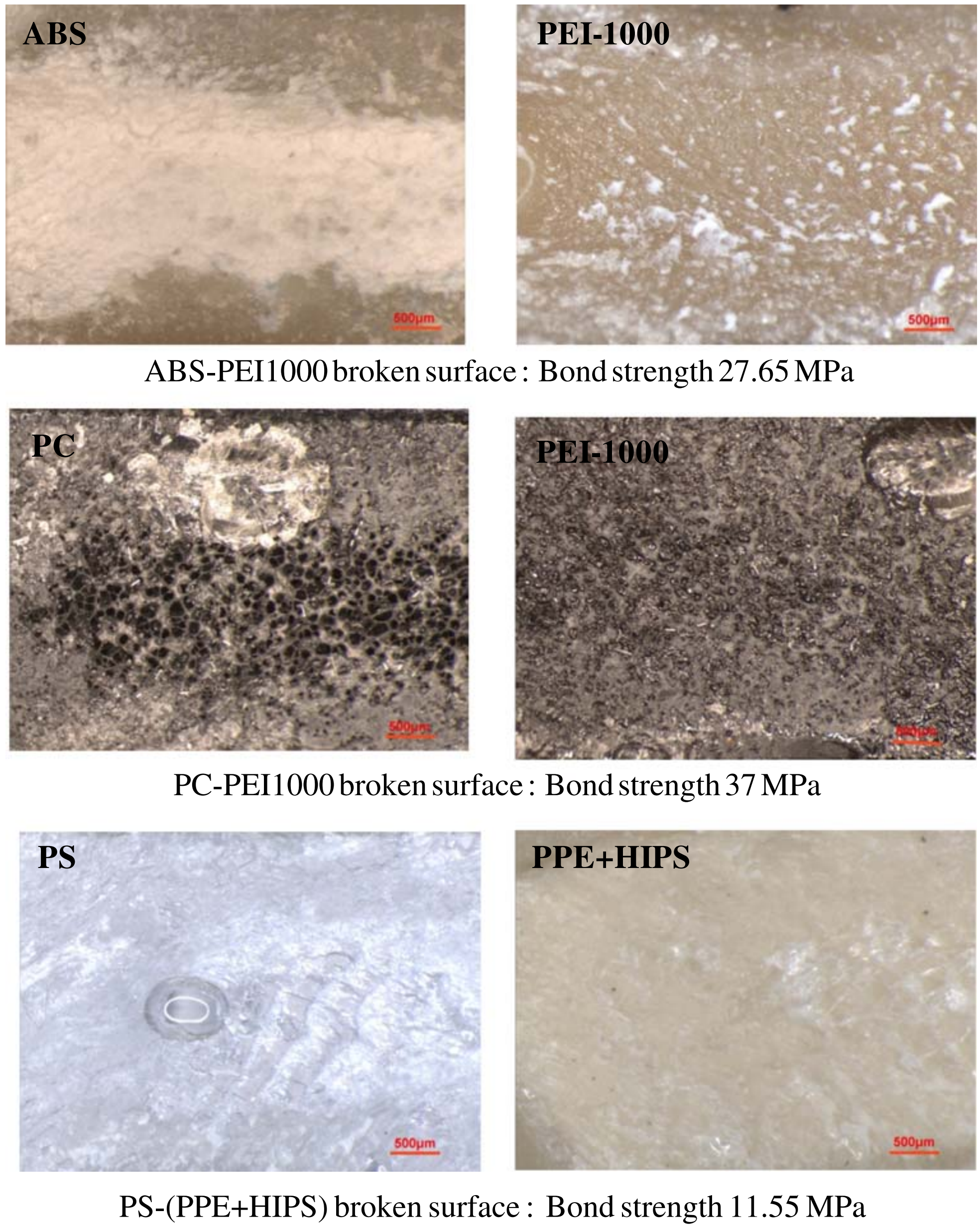


Fig. 11 Broken interface of 2K moulded plastic parts

factors demands special attention because the post-moulded parts undergo special environmental conditions in the metallization process. The moisture and corrosion stress simulate the metallization environment and temperature test simulate basically the service condition. The results from this investigation provide comprehensive information and recommendations concerning the loss of polymer–polymer bond strength during the metallization process. Three combinations of materials were selected for the environmental tests: ABS-PC, PC-PEI and PEI-PEEK. In Table 4, summarised test conditions are shown.

Figure 13a shows the effects of different test conditions on different materials pairs with respect to the references. Figure 13b indicates the comparative effects of different test environments on different material pairs. All the combinations are adversely affected by the boiling water and the percentage reduction in bond strength is almost linear with the weight percentage of water absorption by the two polymers which means the higher the water absorption by the two polymers, the higher the reduction in bond strength (Fig. 14a). The most interesting result was observed in the temperature test. Under this test, ABS-PC and PC-PEI were weakened but PEI-PEEK combination gained bond strength. To understand this abnormality, more tests were done with PEI-PEEK.

Figure 14b shows the temperature test results from PEI-PEEK combination. A longer time at elevated temperature increased the adhesion even further. The material pair was strongest in 160°C for 1 h. In 240°C for 1 h, the bond strength decreased drastically. The possible reason for

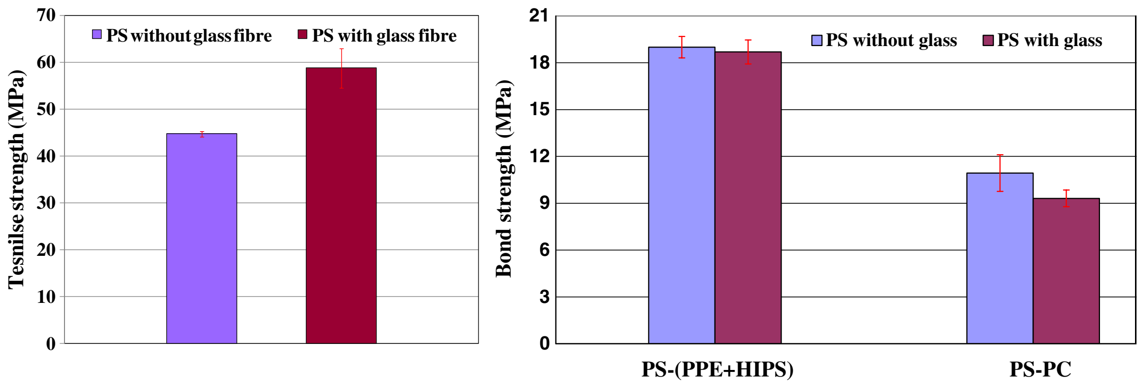


Fig. 12 Effects of glass fibre on polymer–polymer adhesion

Table 4 Recipe for different environmental tests

Test name	Recipe
Moisture test	Boiling of 2 k samples in water (100°C) for 10 min
Corrosion test	2 k samples in 25% (v/v) H_2SO_4 solutions for 30 min
Temperature test	Samples are at -18°C for 72 hr and in +80°C for 30 min

increased bond strength of PEI-PEEK under temperature treatment could be in the fact of co-crystallisation at the interface between PEI and PEEK. PEEK is a semi-crystalline material and it has a glass transition temperature of 143°C. A temperature annealing slightly above its T_g helps PEEK to form more crystals and make a stronger bond with PEI. On the other hand, at 240°C, PEI starts softening and loosens the bond with PEEK.

7 Conclusion

The adhesion mechanism of two-component moulded polymer parts is complex and is a combination of many effects or phenomenon. The conclusion from these experiments can be summarised as follows.

1. In two-component moulding, the bond between two polymers is usually weaker than the individual strength of the any of the materials in the pair. In the best case, about 60% of the strength of the weakest material in the pair was found.

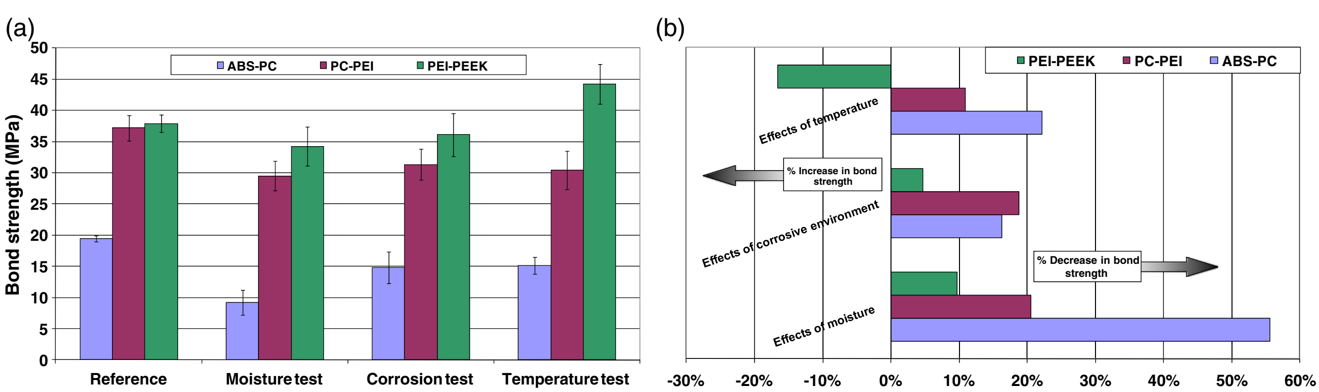


Fig. 13 **a** Effects of environmental factors on bond strength. **b** Effects expressed as a percentage of the original bond strength

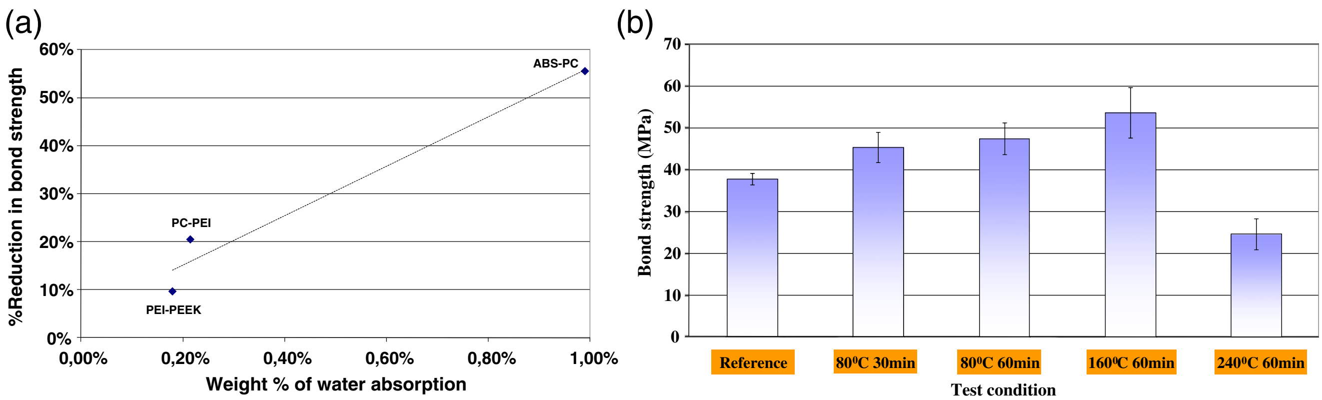


Fig. 14 **a** Effects of water absorption on bond strength. **b** Effects of temperature annealing on PEI-PEEK samples

2. Among the injection moulding parameters, mould and melt temperature were found to be the most significant for the bond strength of two-shot moulded parts. Injection speed, pressure and holding pressure had smaller effect on the bond strength of two-component polymer parts.
3. The interface temperature during the second-shot moulding is vital for the adhesion of two polymers. If the two polymers cannot melt together, the adhesion is usually poor. The thermal and heat transfer properties of the two materials affect the interface temperature and bonding.
4. Interface roughness increases mechanical locking and localised melted zone, so the materials can have a higher rate of entanglement and higher strength. Glass fibre in the first-shot inlay part reveals no significant influence on the bond strength with a second polymer.
5. Environment factors like moisture, corrosive environment and thermal history affects the bond strength of the post-moulded parts. Thermal annealing increase the bond strength between polymers when at least one of the polymers in the pair is semi-crystalline. The annealing slightly above the glass transition temperature helps co-crystallisation of two polymers at the interface and makes the bond stronger.

Acknowledgement This paper reports work undertaken in the context of the project “COTECH-Converging Technologies for Micro System Manufacturing”. COTECH is a Large-scale Collaborative Project supported by the European Commission in the 7th Framework Programme (CP-IP 214491-2). Furthermore, the Danish Innovation-konsortium “POLYMETAL” (contract no. 61568) is acknowledged.

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